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Study of Chelyabinsk LL5 Meteorite Fragment with a Light Lithology and Its Fusion Crust Using Mössbauer Spectroscopy with a High Velocity Resolution

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Abstract. Study of Chelyabinsk LL5 ordinary chondrite fragment with a light lithology and its fusion crust, fallen on February 15, 2013, in Russian Federation, was carried out using Mössbauer spectroscopy with a high velocity resolution. The Mössbauer spectra of the internal matter and fusion crust were fitted and all components were related to iron-bearing phases such as olivine, pyroxene, troilite, Fe-Ni-Co alloy, and chromite in the internal matter and olivine, pyroxene, troilite, Fe-Ni-Co alloy, and magnesioferrite in the fusion crust. A comparison of the content of different phases in the internal matter and in the fusion crust of this fragment showed that ferric compounds resulted from olivine, pyroxene, and troilite combustion in the atmosphere.

Keywords: Chelyabinsk LL5 ordinary chondrite, fragment with a light lithology, fusion crust, Mössbauer spectroscopy with a high velocity resolution.
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INTRODUCTION

Ordinary chondrites consist of various iron-bearing phases such as olivine $(Fe, Mg)_2SiO_4$, pyroxene $(Fe, Mg, Ca)SiO_3$, troilite (FeS), metal grains (Fe–Ni–Co alloys), chromite ($FeCr_2O_4$), daubréelite ($FeCr_2S_4$), *etc.*, as well as its weathering products in the form of ferric compounds. Ordinary chondrites contain 19–28 wt% of total Fe, including 10–18 wt% of Fe in silicates, 4–6 wt% of Fe in troilite, and 2–17 wt% of Fe in metal grains containing α-Fe(Ni, Co) and γ-Fe(Ni, Co) phases [1]. They are related to three groups: H, L, and LL. These groups are characterized by different total iron content and different metal content: H – high iron content, L – low iron content, and LL – low iron and low metal content. The content of total iron in H, L, and LL groups of ordinary chondrites is ranged from 25 to 28 wt%, from 20 to 25 wt%, and from 19 to 22 wt%, respectively, while the content of metal in these groups is of 15–19 wt%, 4–10 wt%, and 1–3 wt%, respectively [2].

A bright fireball was seen by numerous residents in the Kurgan, Tyumen, Sverdlovsk, and Chelyabinsk regions of Russian Federation in the morning on February 15, 2013. A lot of stone fragments fell in the Chelyabinsk region. These meteorite fragments were further classified as ordinary chondrite LL5 group, shock stage S4, weathering W0 and named Chelyabinsk (Meteoritical Bulletin No. 102). The Meteoritical expedition of the Ural Federal University immediately collected fragments of this meteorite. These fragments appeared to be different in size ranging from 1–2 mm up to 15–17 cm and demonstrated different lithology. The first study of the Chelyabinsk LL5 fragment with a light lithology using Mössbauer spectroscopy with a high velocity resolution showed the presence of olivine, pyroxene, troilite, and metal in the sample with the absence of any ferric compounds due to the absence of fragment

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weathering [3]. The fragment also showed a very low metal content (of ~1.7%). The obtained Mössbauer parameters were in agreement with previously published data for ordinary chondrites (see, for instance, [4–13]). In the present work, we consider the results of the study of another fragment of Chelyabinsk LL5 meteorite with a light lithology and its fusion crust carried out using Mössbauer spectroscopy with a high velocity resolution.

EXPERIMENTAL DETAILS

The fragment of Chelyabinsk LL5 meteorite with a light lithology studied in this work is shown in Fig. 1. The thickness of fusion crust in this fragment was about 0.10–0.15 mm. The slice of this fragment was prepared for metallography and scanning electron microscopy (SEM). Then, the powdered sample of the internal matter was prepared from this slice for further X-ray diffraction (XRD) study and Mössbauer spectroscopy. Additionally, the sample of the fusion crust was prepared as a powder. Both powdered samples for Mössbauer spectroscopy were glued on Al foil free from iron with a surface thickness of less than 10 mg Fe/cm^2.

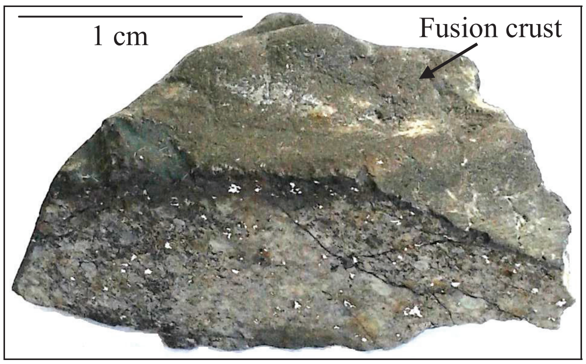

FIGURE 1. Fragment of Chelyabinsk LL5 ordinary chondrite with a light lithology.

The slice of meteorite fragment was analyzed by means of optical microscopy with an Axiovert 40 MAT and a scanning electron microscopy using ΣIGMA VP microscope (Carl Zeiss) with energy dispersion spectroscopy (EDS) for chemical analysis. The phase analysis of meteorite samples was performed by X-ray powder diffraction employing scanning rate of 0.026 °/s in 2Θ ranges from 14° to 80° for the internal matter and from 20° to 100° for the fusion crust using an X'Pert PRO MRD diffractometer with Cu K_{α} radiation.

Mössbauer spectra with a high velocity resolution were measured using an automated precision Mössbauer spectrometric system based on a SM–2201 spectrometer with a high velocity resolution and liquid nitrogen cryostat with moving absorber. The SM–2201 spectrometer operates with the saw-tooth shape velocity reference signal formed by digital-analogue convertor using quantification with 4096 steps and spectra registration in 4096 channels. Details and characteristics of this spectrometer and the system were given in [14–16]. The ~1.0×10^9 Bq ^{57}Co(Rh) source (Ritverc GmbH, St. Petersburg) was used at room temperature. The Mössbauer spectra of meteorite samples were measured in the transmission geometry with moving absorber in the cryostat at 295 K and recorded in 4096 channels. Then, these spectra were converted in 1024 channels by consequent summation of four neighboring channels to increase the signal-to-noise ratio especially for the minor spectral components. Statistics was ~5.0×10^5 and ~7.3×10^5 counts per channel and the signal-to-noise ratio was 68 and 94 for the spectra of the internal meteorite matter and the fusion crust, respectively. The Mössbauer spectra were computer fitted with the least squares procedure using an UNIVEM-MS program with a Lorentzian line shape. The spectral parameters such as isomer shift, δ, quadrupole splitting (quadrupole shift for magnetically split subspectra), ΔE_Q, magnetic hyperfine field, H_{eff}, line width, Γ, relative subspectrum area, S, and statistical criterion, χ^2, were determined. Magnetic sextets were fitted using the sextet peaks area ratio as $S_{1,6}$:$S_{2,5}$:$S_{3,4}$ = 3:2:1. Instrumental (systematic) error for the velocity scale or systematic error was ±0.5 channel for each spectral point and ±1 channel for hyperfine parameters in the case of measurements using the SM-2201 spectrometer. It should be noted that this spectrometer's characteristics determined an integral velocity error (total mechanical and electronics systematic and random errors) which was several times less than a half of channel value in mm/s during spectra measurements using 4096 channels [14]. Velocity resolution (velocity per one channel) was ~0.015 mm/s per channel and ~0.019 mm/s per channel for the 1024 channels spectra of the internal meteorite matter and the fusion crust, respectively. The relative error for S did not exceed 10%. If an error calculated with the fitting procedure (fitting error) for these parameters exceeded the instrumental (systematic) error we used the larger error instead. Criteria of the best fit involved differential spectrum, χ^2, and physical meaning of parameters. Values of δ are given relative to α-Fe at 295 K.

RESULTS AND DISCUSSIONS

Images of Chelyabinsk LL5 fragment with a light lithology obtained using optical and scanning electron microscopy are shown in Fig. 2. These images demonstrate the presence of metal grains and troilite inclusions in silicate matrix. The results of chemical analysis obtained using EDS showed that metal grains contained α-Fe(Ni, Co) and γ-Fe(Ni, Co) phases. Concentration of Ni in the α-phase was in the range from ~4 to ~5 at% and that of Co was in the range from ~2.5 to ~3.0 at%. Concentration of Ni in the γ-phase was in the range from ~33 to ~44 at% and that of Co was in the range from ~0.0 to ~1.5 at%. The troilite inclusions contained ~51–52 at% of S and ~49–48 at% of Fe.

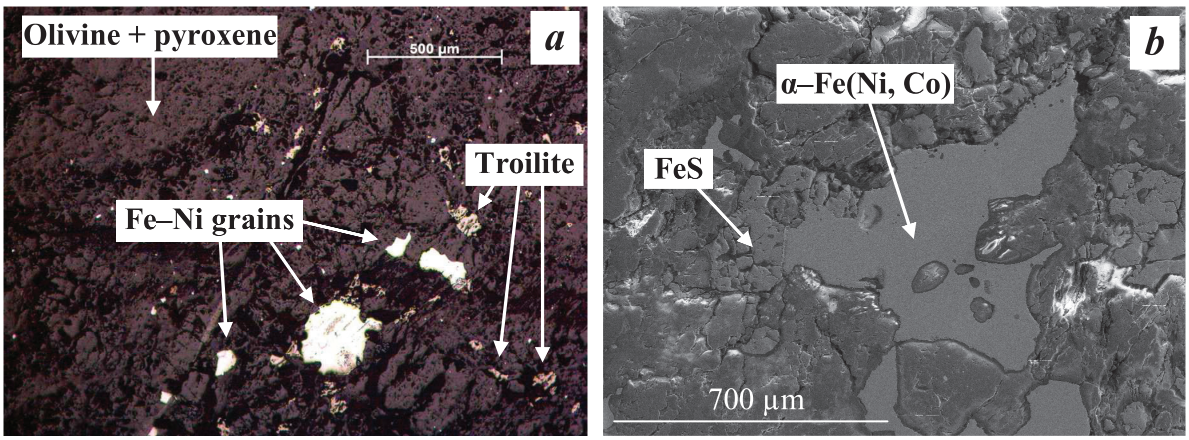

FIGURE 2. Microphotographs of the Chelyabinsk LL5 meteorite fragment obtained using (a) optical microscopy and (b) scanning electron microscopy.

XRD patterns of the internal matter and the fusion crust of Chelyabinsk LL5 meteorite fragment are shown in Fig. 3. The obtained data for the internal matter indicate the presence of olivine (~66 wt%), pyroxene (~29 wt%) and troilite (~5 wt%) phases while metal phase content is very small to be distinguished well. The fusion crust XRD pattern demonstrates also the presence of olivine (~50 wt%), pyroxene (~27 wt%), troilite (~4 wt%) phases, Fe-Ni-Co alloy (~1 wt%) and an additional phase related to magnesioferrite ($MgFe_2O_4$) (~18 wt%).

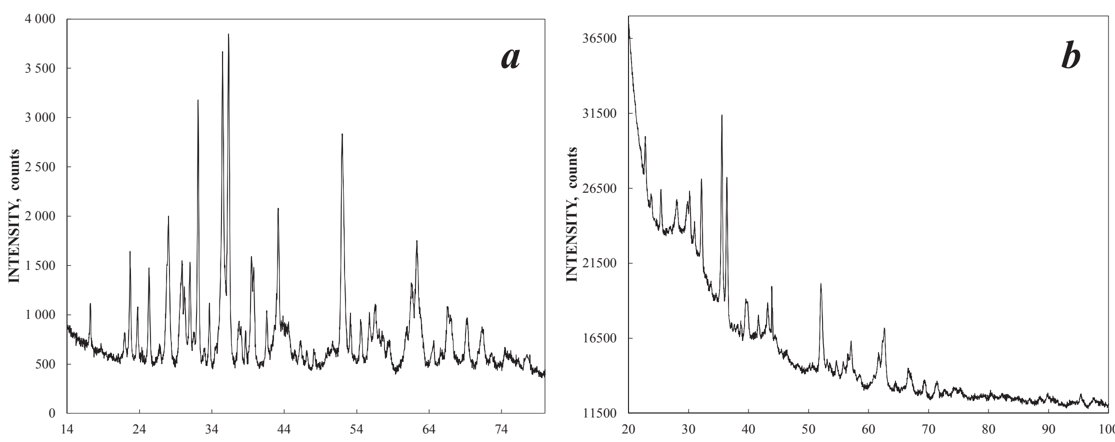

FIGURE 3. X-ray diffraction patterns for (a) the internal matter and (b) the fusion crust of the Chelyabinsk LL5 ordinary chondrite fragment.

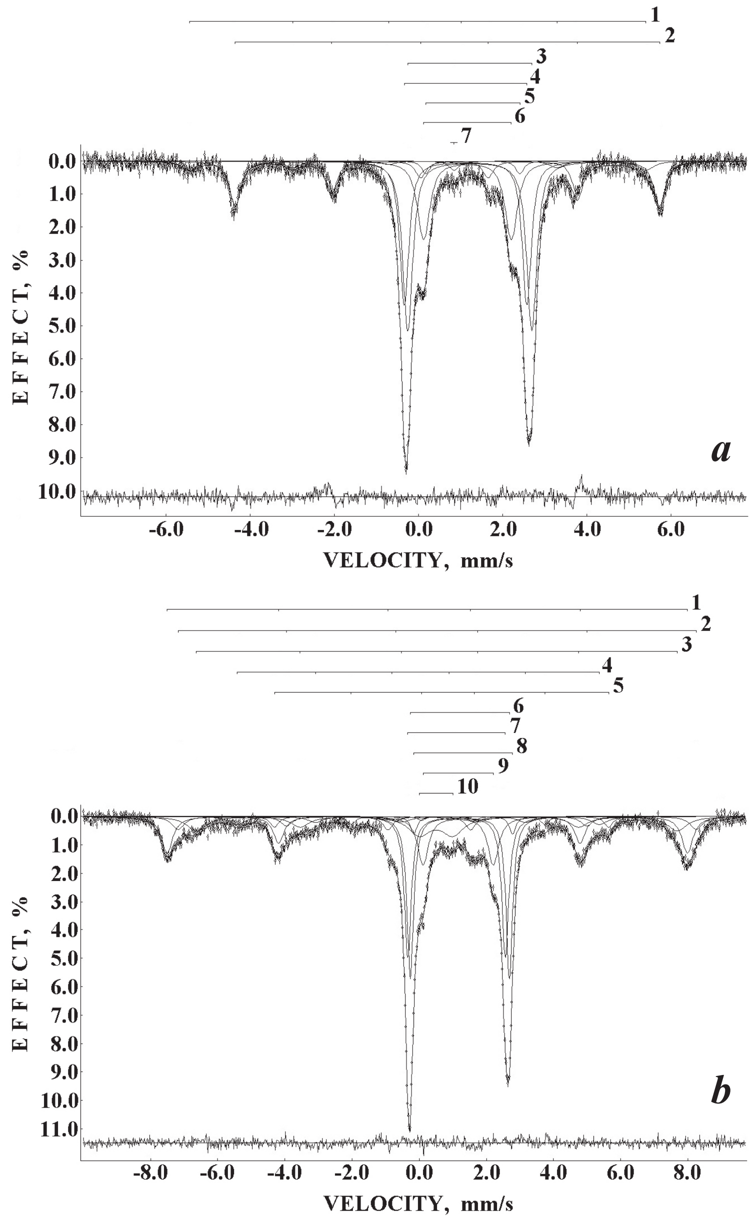

FIGURE 4. The Mössbauer spectra of (a) the internal matter and (b) the fusion crust of the Chelyabinsk LL5 ordinary chondrite fragment with a light lithology. Indicated components are the results of the better fit. Differential spectra are shown below.

The Mössbauer spectra of the internal matter and the fusion crust of the Chelyabinsk LL5 meteorite fragment are shown in Fig. 4. The Mössbauer spectrum of the internal matter is similar to other spectra of ordinary chondrites (see [3–13]). This spectrum was fitted well using two magnetic sextets, four quadrupole doublets and one paramagnetic singlet. Parameters are given in Table 1. The Mössbauer spectrum of the fusion crust was fitted using five magnetic sextets and five quadrupole doublets parameters of which are given in Table 1. Two magnetic sextets in the spectrum of the internal matter were related to Fe-Ni-Co alloy (component 1) and FeS (component 2). Quadrupole doublets 3 and 4 were related to the ^{57}Fe in crystallographically non-equivalent sites denoted as M1 and M2 in olivine while quadrupole doublets 5 and 6 were related to the ^{57}Fe in crystallographically non-equivalent sites denoted as M1 and M2 in pyroxene (see [8]). It was interesting to observe an additional minor paramagnetic singlet (component 7) the δ value of which was close to that for chromite $FeCr_2O_4$ (see, for instance, [17] in which the authors fitted the room-temperature single peak using a quadrupole doublet with ΔE_Q value of 0.06 mm/s and δ value of 0.92 mm/s, and [18] in which the authors fitted the spectrum of pure $FeCr_2O_4$ as a single peak with δ value of 0.92 mm/s). It should be noted that at room temperature, the paramagnetic singlet can be observed for daubréelite ($FeCr_2S_4$), another mineral found in various meteorites containing troilite, however, its δ value (see [19]) was slightly different from that of chromite. Therefore, we assigned component 7 with chromite. In the component 1 related to Fe-Ni-Co alloy, the Γ value was broad that may be a result of contribution from α- and γ-phases with variations in Ni and Co concentration the subspectra of which cannot be resolved in this component due to a very poor signal-to-noise ratio resulting from a small amount of metal in ordinary chondrites from the LL group.

TABLE 1. Mössbauer parameters of the internal matter and the fusion crust of the Chelyabinsk LL5 ordinary chondrite fragment with a light lithology. T = 295 K.

Sample	Γ, mm/s	δ, mm/s	ΔE_Q, mm/s	H_{eff}, kOe	S, %	Component[a]
Internal matter	0.455 ± 0.065	0.067 ± 0.020	– 0.170 ± 0.038	336.9 ± 1.4	5.2	Fe-Ni-Co alloy (1)
	0.330 ± 0.032	0.771 ± 0.016	– 0.169 ± 0.016	313.1 ± 0.5	19.0	Troilite (2)
	0.287 ± 0.032	1.219 ± 0.016	2.951 ± 0.016	–	31.1	Olivine M1 (3)
	0.263 ± 0.032	1.120 ± 0.016	2.913 ± 0.016	–	24.1[b]	Olivine M2 (4)
	0.233 ± 0.032	1.294 ± 0.024	2.233 ± 0.048	–	2.0	Pyroxene M1 (5)
	0.357 ± 0.032	1.165 ± 0.016	2.084 ± 0.016	–	17.7	Pyroxene M2 (6)
	0.294 ± 0.082	0.846 ± 0.027	–	–	0.9	Chromite (7)
Fusion crust	0.400 ± 0.040	0.271 ± 0.020	– 0.058 ± 0.020	481.0 ± 0.6	15.7	$MgFe_2O_4$ (A) (1)
	0.453 ± 0.084	0.528 ± 0.029	0.015 ± 0.024	479.0 ± 1.3	6.3	$MgFe_2O_4$ [B] (2)
	0.704 ± 0.063	0.562 ± 0.020	– 0.079 ± 0.028	444.6 ± 1.8	10.8	$MgFe_2O_4$ [B] (3)
	0.566 ± 0.073	0.005 ± 0.023	0.069 ± 0.042	335.1 ± 1.5	4.9	Fe-Ni-Co alloy (4)
	0.309 ± 0.040	0.760 ± 0.020	– 0.191 ± 0.023	308.9 ± 0.9	3.6	Troilite (5)
	0.233 ± 0.040	1.210[b]	2.949 ± 0.020	–	20.3	Olivine M1 (6)
	0.248 ± 0.040	1.113 ± 0.020	2.903 ± 0.020	–	18.8	Olivine M2 (7)
	0.233 ± 0.040	1.309 ± 0.020	2.939 ± 0.021	–	2.2	Pyroxene M1 (8)
	0.390 ± 0.040	1.160 ± 0.020	2.091 ± 0.020	–	10.1	Pyroxene M2 (9)
	0.776 ± 0.063	0.502 ± 0.026	0.993 ± 0.047	–	7.4	Paramagnetic $MgFe_2O_4$ (10)

[a] Numbers in parenthesis correspond to components numbers in Fig. 4.
[b] Fixed parameters.

It is possible to compare the results of Mössbauer spectroscopy of two different fragments of the Chelyabinsk LL5 meteorite with a light lithology measured previously (fragment 1) [3] and in the present work (fragment 1a). If we suggest that the f-factor is similar for all the phases, we can evaluate the relative content of each phase in the studied samples. In this case, the metal content in fragment 1a was found larger than that in fragment 1 (5.2% and 1.7%, respectively). The larger content of troilite was also found in fragment 1a in comparison with that in fragment 1 (19.0% and 12.7%, respectively). The content of olivine was the same within the error in both fragments (55.2% and 57.6%, respectively), while the content of pyroxene was found larger in fragment 1 (28.0%) in comparison with that in fragment 1a (19.7%). Additionally, we revealed 0.9% of chromite in fragment 1a. These differences may be a result of some variation of phases' concentrations through a big stone which was destroyed in the atmosphere or a result of a breccia structure of this meteorite resulting from various collisions of parent bodies in the space forming Chelyabinsk LL5 meteorite from parts of these different bodies.

The Mössbauer spectrum of the fusion crust showed the presence of new components such as magnetic sextets (components 1–3) and quadrupole doublet with parameters corresponding to ferric compound (component 10). Based on the results of XRD study, these components were related to magnesioferrite which may be a product of combustion of silicate phases. Different occupations of tetrahedral (A) and octahedral [B] positions with Fe and Mg may be a reason of different microenvironments of the ^{57}Fe nuclei in both (A) and [B] positions leading to several magnetic sextets (for instance, a correlation of the probabilities of different numbers of Ni atoms in the ^{57}Fe nuclei local microenvironment with different number of magnetic sextets related to the ^{57}Fe in both (A) and [B] sites was found in $NiFe_2O_4$ using Mössbauer spectroscopy with a high velocity resolution [20, 21]). Taking into account the difference between δ values for magnetic sextets 1 and both 2 and 3, we related component 1 to the ^{57}Fe in (A) sites while components 2 and 3 were related to the ^{57}Fe in [B] sites (see Table 1). Component 10 (quadrupole doublet) was also related to $MgFe_2O_4$ particles with a very small size (XRD results indicated that there is a size variation for magnesioferrite crystals with the smallest size of about several nanometers which were distributed in the glass-like fusion crust after fast cooling in the atmosphere). These results are in agreement with the recent Mössbauer study of nanostructured $MgFe_2O_4$ [22]. The content of magnesioferrite in the fusion crust was evaluated to be about 40%, the metal content was the same as in the internal matter while troilite content was found to be about five times decreased. The content of both olivine (39.1%) and pyroxene (12.3%) in the fusion crust was also smaller than that in the internal matter (55.2% and 19.7%, respectively). Using these results, we can suppose that the fusion crust formation was realized mainly by combustion of olivine, pyroxene, and troilite leading to appearance of ferric compound in the form of magnesioferrite particles with different size resulting in magnetic and superparamagnetic states of $MgFe_2O_4$ at room temperature. These particles were distributed in the fusion crust after cooling. We should suppose that sulfur from troilite was removed from the fusion crust after combustion or did not bind iron containing compounds in sufficient quantities to be detected by Mössbauer spectroscopy.

CONCLUSIONS

Study of the second fragment with a light lithology of the Chelyabinsk LL5 ordinary chondrite was carried out using Mössbauer spectroscopy with a high velocity resolution. The phase composition determined in this fragment was different from that in the first fragment also with a light lithology studied earlier in [3]. This difference may be a result of the breccia structure of the Chelyabinsk LL5 meteorite which was formed by collisions of different parent bodies in the space. The fusion crust of this fragment contains a large amount of ferric iron in the form of magnesioferrite which was formed by olivine, pyroxene, and troilite combustion in the atmosphere. Therefore, the content of silicates and troilite was found decreased in comparison with that in the internal matter of this fragment.

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